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Atmospheric Scavenging Processes

by

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1 Introduction

As was discussed in lecture (1), there are a number of processes which release and generate aerosol in the atmosphere. The very much higher concentration of some particulates downwind of areas of urban activity confirms that there are substantial anthropogenic sources. Indeed man deliberately, if somewhat unconsciously, uses the atmosphere as a sink for gaseous and particulate waste products. The fact that the concentration of most, if not all, of these does not increase monotonically with time demonstrates that there must be one or more processes which destroy or remove them from the atmosphere. In fact almost all aerosol is removed by being transported to and deposited at the earth's surface.

Removal mechanisms for particulates are classified as 'wet' or 'dry' depending upon whether the removal is via precipitation (rain, snow, settling fog drops, etc.) or by deposition in the dry state. This discussion will concentrate on the essential features of a number of microphysical processes which contribute to the 'wet' process, as a preliminary to reporting the present state of understanding of their net effect. Last year, in the context of the global cycles of various trace chemical species, Dr Adrian Tuck described some of the large scale characteristics and effects of these processes.

2 Physical processes

2.1 Diffusion

The macroscopic concept of a diffusion coefficient is that of the constant of proportionality between the flux of some property of a fluid and the concentration gradient which drives it.

Thus $$\underline{j} = -D\nabla n \qquad (1)$$

Application of the equation of continuity for the diffusing substance,

$$\frac{\partial n}{\partial t} + \nabla \cdot \underline{j} = 0$$

$$\frac{\partial n}{\partial t} = D\nabla^2 n \qquad (2)$$

leads to the so-called 'diffusion equation'

For the case of an isolated sphere of radius r having perfect accomodation at its surface, so that $n(r) = 0$; in an environment containing n_∞ of aerosol per unit volume at large distances from it, the flux equation (1) has a steady state solution.

$$j_{radial} = 4\pi r D n_\infty \quad \text{and} \quad n(x) = n_\infty \left(1 - \frac{r}{x}\right)$$

where x is the radial distance from the centre of the sphere.

When diffusion is viewed as a consequence of Brownian motion, ie. as a microscopic concept, then the diffusion coeffficient has a microphysical interpretation:

$$D = \frac{kT}{6\pi a \eta} \quad \text{for } a > \lambda$$

$$D = \frac{kT\lambda}{6\pi a^2 \eta} \quad \text{for } a \leq \lambda$$

where a is the radius of the diffusing particle

λ is the molecular mean free path,

η is the dynamic viscosity.

These expressions highlight the fact that diffusion is maximised for small particles in a fluid of low viscosity and high temperature. The dependence on λ reflects the need to correct for the use of a continuum concept of viscosity when the mean free path of the molecules is greater than the size of the aerosol experiencing the effect of viscous drag. The ratio is known as Knudsen's number, Kn. $\lambda \simeq 0.1\,\mu m$ at STP,

so that aerosol physics is concerned with both high and low values of Kn.

Applying the above results to a distribution of particles of sufficiently low number density for their diffusion fields to be independent of one another (this is a good approximation in cloud and precipitation where the average drop separation $\gg$ 100 drop radii): If there are $n(a)\,\Delta a$ aerosol particles in the size range $a \rightarrow a+\Delta a$ while there are $N(r)\,\Delta r$ drops which act as collecters then the rate of change of concentration of the former resulting from diffusion to the latter is

$$-4\pi r D(a) N(r) n(a) \Delta r \Delta a$$

and integrating over all sizes of collecting drop

$$\frac{dn(a)}{dt} = -4\pi D(a) n(a) \int_0^\infty r N(r) dr \qquad -(3)$$

The integral can be evaluated but for simplicity is often written as $N\bar{r}$, where N is the number density of all drops and $\bar{r}$ is their average radius.

Thus $$\frac{1}{n(a)} \frac{dn(a)}{dt} = -4\pi \bar{r} N D(a) = -\Lambda_{diff} \qquad -(4)$$

where Λ_{diff} is the so called scavenging coefficient for diffusion.

Thus $$n(a,t) = n(a,0) \exp(-\Lambda t) \qquad -(5)$$

A more illustrative definition of Λ can be achieved by recalling that the liquid water content of cloud, ω, can be expressed as:

$$\omega = \frac{4}{3}\pi \bar{r}^3 N \rho_L$$ where ρ_L is the density of water

Thus $$\Lambda = \left(\frac{48\pi^2 \omega N^2}{\rho_L}\right)^{1/3} D$$

Maritime cloud can be characterised by $N \sim 100\ cm^{-3}$, $\omega \sim 0.3\ g\ m^{-3}$ and continental cloud by $N \sim 1000\ cm^{-3}$, $\omega \sim 0.3\ g\ m^{-3}$.

Then: $\Lambda_{MAR} = 1.12 D\ sec^{-1}$ and $\Lambda_{CONT} = 5.22 D\ sec^{-1}$.

Figure 2 demonstrates the e - folding time ($1/\Lambda$) for aerosol of radius a in such clouds. Efficient capture of particles below 0.01 μm, especially in continental clouds and fog, with no discernable capture of 0.1 μm particles, is predicted.

When there is an appreciable airflow relative to the collecting drop, the diffusion equation becomes

$$\frac{\partial n}{\partial t} = D\nabla^2 n - v.\nabla n \qquad \text{— (6)}$$

where v is the flow field relative to the drop.

The ratio of diffusive and convective transport is

$$\frac{v.\nabla n}{D\nabla^2 n} \simeq \frac{2V_\infty r}{D} = Pe$$ - the Peclet number.

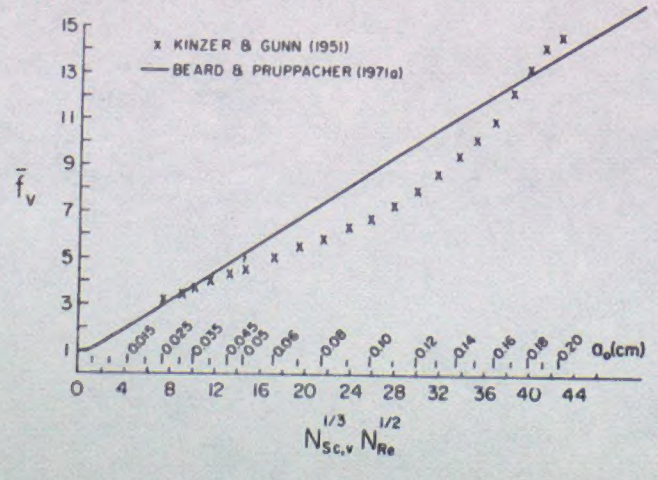
The flow field characteristics are a function of Reynolds number,

$$Re = \frac{\rho r V_\infty}{\eta}$$ where ρ is the air density.

Apparently the effects of the flow field on diffusion is a function of these two numbers only. It is common in cloud physics to take account of the effect of ventilation by a factor f_v

$$f_v = A + B\, Re^{2/3} Pe^{1/3}$$

A and B are weak functions of R_e but for most purposes $A \simeq 1$ and $B \sim 0.3$ will suffice.



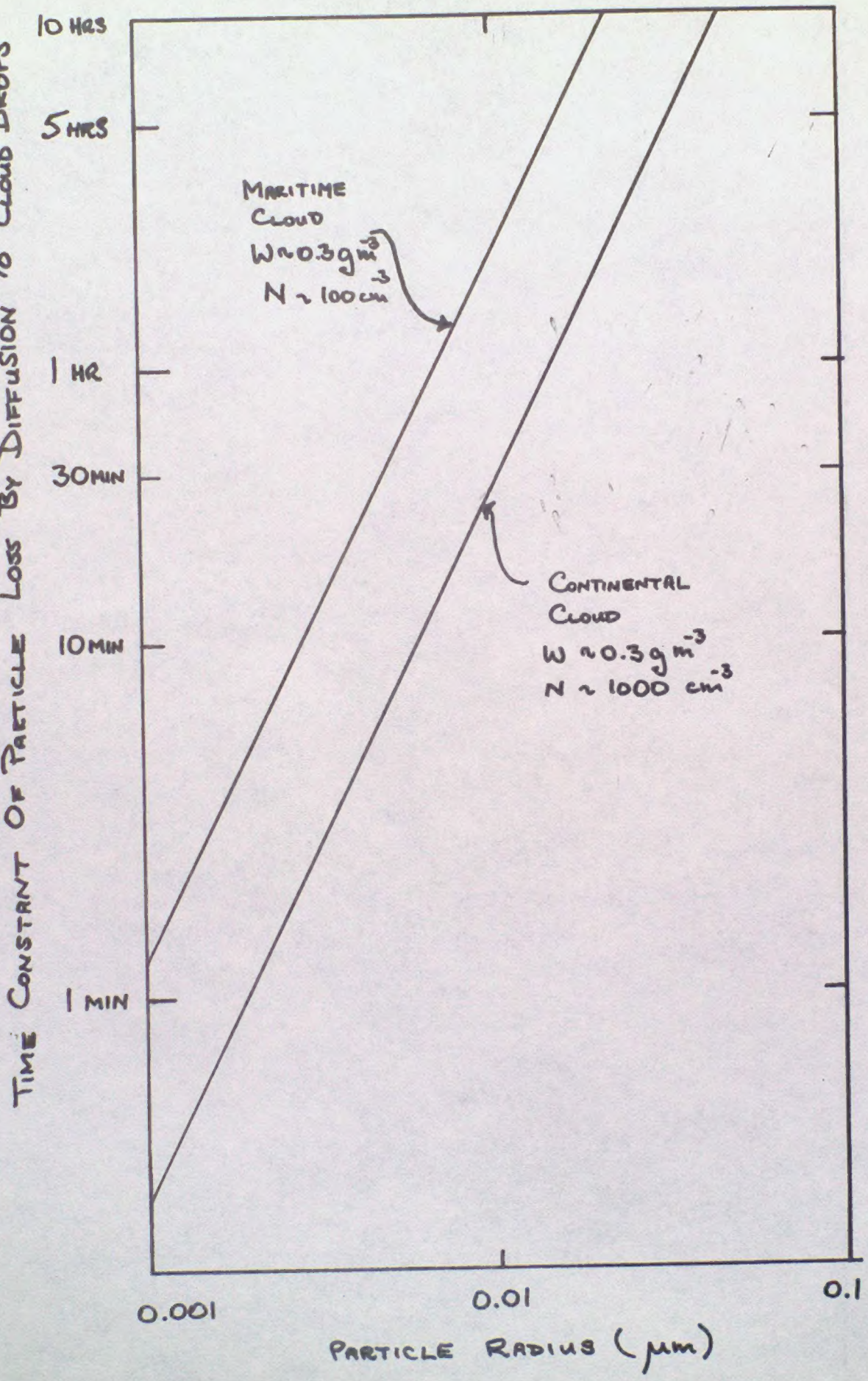

10 HRS
5 HRS
1 HR
30 MIN
10 MIN
1 MIN
TIME CONSTANT OF PARTICLE LOSS BY DIFFUSION TO CLOUD DROPS
MARITIME CLOUD
W ~ 0.3 g m⁻³
N ~ 100 cm⁻³
CONTINENTAL CLOUD
W ~ 0.3 g m⁻³
N ~ 1000 cm⁻³
0.001
0.01
0.1
PARTICLE RADIUS (μm)

Fig. 2

The predictions of Beard and Pruppacher (1971) are shown above in comparison with the experimental results of Kinzer and Gunn(1951).

These ventilation enhancements have only a small effect on the results of fig 2.

Thus $\Lambda = 4\pi\bar{r}ND\,f_v$ for a moving drop where $f_v \leq 10$

2.2 Phoretic effects

2.2.1 Diffusiophoresis

Aerosol particles suspended in a non-uniform but isothermal mixture of gases, move in response to concentration gradients. The aerosol move, in general, in the direction of the diffusive flux of the heavier gas component. Thus the diffusiophoretic flux is expected to be away from an evaporating drop but towards one that is experiencing condensational growth.

The drift velocity due to a water vapour gradient ∇n_w is given by

$$U_D \propto \frac{\sqrt{M_w}}{n_A\sqrt{M_A}} D_w \nabla n_w \qquad \text{— (7)}$$

where Dw is the diffusion coefficient for water vapour
M_w, M_A are the effective molecular weights of water and air
n_w, n_A are the number densities of these molecules

The experimental work of Goldsmith et al (1963) and Goldsmith and May (1966) confirms the form of this relationship, which numerically can be written as:

$$U_D = 1.9\,10^{-7}\,kT\,\frac{dn_w}{dr}.$$

In an environment of n aerosol and N drops per unit volume the rate of removal by diffusiophoresis is

$$\frac{dn}{dt} = -1.9\,10^{-7}\,kT\,Nn\,4\pi r^2\,\frac{dn_w}{dr}$$

where $4\pi r^2 D M_w \frac{dn_w}{dt} = \frac{d\omega}{dt}$ where ω is the liquid water content of the cloud

Hence $$\Lambda = -\frac{1}{n}\frac{dn}{dt} = \frac{1.9\,10^{-7}\,kT}{DM_w}\frac{d\omega}{dt}$$

Although this may be large locally when $d\omega/dt$ is large, integration through the formation of a cloud yields

$n = n_o \exp(-10^3\omega)$ and as $\omega \leq 10^{-6}$ diffusiophoresis is not a very effective collection mechanism in total.

2.2.2 Thermophoresis

Thermophoresis is the name given to the motion of particles caused by the non-uniform temperature of a gas. It may be thought of as a result of air molecules giving a greater net impulse on the warm side of the particle than on the cold side, thereby driving it in the direction of the temperature gradient. Thus, this effect is expected to lead to net motion of aerosol towards an evaporating drop. The drift velocity has the form

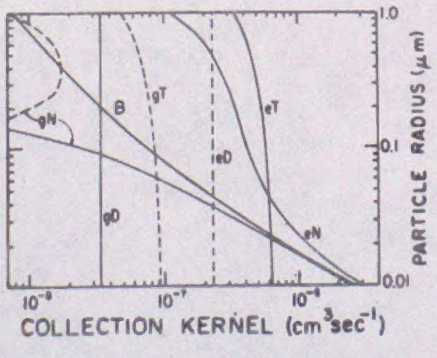
$$V_t \propto \frac{k_T}{p} \nabla T$$

where k_T is the thermal conductivity of the air
p is the ambient pressure.

Clearly thermophoresis cannot be thought of as acting in isolation from diffusiophoresis. Evidently there is coupling between the molecular flux of water Fw and the heat flux F_t since

$$F_t = L F_\omega \ , \qquad \text{L being the latent heat.}$$

Young (1974) has carried out numerical evaluation of the simultaneous effects of diffusion, thermo-and diffusiophoresis for various atmospheric conditions. His results for the case of a water drop of 10μm radius evaporating at 98% relative humidity, 600mb and -5°C; and for a drop growing at a supersaturation of 0.3%, 600mb and -5°C are displayed below:



Collection kernel ($cm^3 sec^{-1}$) for a water drop of $a = 10\,\mu m$ in air of 600 mb and −5 °C, collecting aerosol particles by Brownian diffusion (B), thermophoresis (T), or diffusiophoresis (D); (N) is net effect; for 0.3% supersaturation (g), 98% relative humidity (e). (From Young, 1974)

The collection kernel , $K(r,a)$ is defined by

$$\Lambda = \int_0^\infty K(r,a)\, N(r)\, dr \qquad \text{—} \qquad (9)$$

In a continental cloud, $N \sim 1000\ cm^{-3}$ $K = 10^{-6}$ corresponds to an e-folding time for collection of the aerosol, radius a, of about 1000 secs; $K = 10^{-7}$ corresponds to a time constant of about 10,000 secs or almost 3 hours. Note that thermophoresis overpowers diffusiophoresis, at least for $a < 1\ \mu m$ where both processes are insensitive to aerosol size, unlike diffusion, which as we have seen is efficient only for small particles. For this reason phoretic effects exceed those for diffusion above $a \sim 0.1\ \mu m$ but remain rather inefficient overall.

2.3 Inertial capture

When a raindrop or large cloud drop falls through an atmosphere containing aerosol some of the latter are collected by the former. However, as was pointed out in lecture (2), not all particles within the swept out volume collide with the drop. Some small particles follow the airflow and avoid collision.

It is possible to quantify this simple physical idea without recourse to complex mathematics. Consider the motion of a particle radius a, along the x axis at velocity u, relative to a fluid of viscosity η. Its equation of motion can be expressed as:

$$m\frac{du}{dt} + 6\pi a\eta u = 0 \qquad — \qquad (10)$$

where the second term is simply the viscous drag on the particle due to its relative velocity u.

Since $u = dx/dt$ the above can be written as:

$$m\frac{du}{dx} = -6\pi a\eta$$

Thus $u = u_0 - \frac{6\pi a\eta}{m}x$ where u_0 is the velocity at x=0.

The particle comes to rest in a distance, x_{stop} where

$$x_{stop} = \frac{mu_0}{6\pi a\eta} = \frac{2\rho_p a^2 u_0}{9\eta} \qquad — \qquad (11)$$

If x_{stop} is small then the particle can be thought of as 'frozen' into the fluid. If the stopping distance is long compared with the characteristic dimension of an obstacle, l say, then the particle tends not to 'make the corner' and a collision results. This simple idea leads to the definition of the Stokes number

$$Stk = \frac{x_{stop}}{l} = \frac{2\rho_p a^2 u_0}{9\eta l} \qquad — \qquad (12)$$

For many purposes (e.g. in aerosol impactors) it is adequate to assume that the collision efficiency is given by:

$$E = Stk \quad \text{if} \quad Stk < 1$$
$$E = 1 \quad \text{if} \quad Stk \geq 1$$

Thus in the case of a particle of radius a by a drop of radius r inertial capture is likely if:

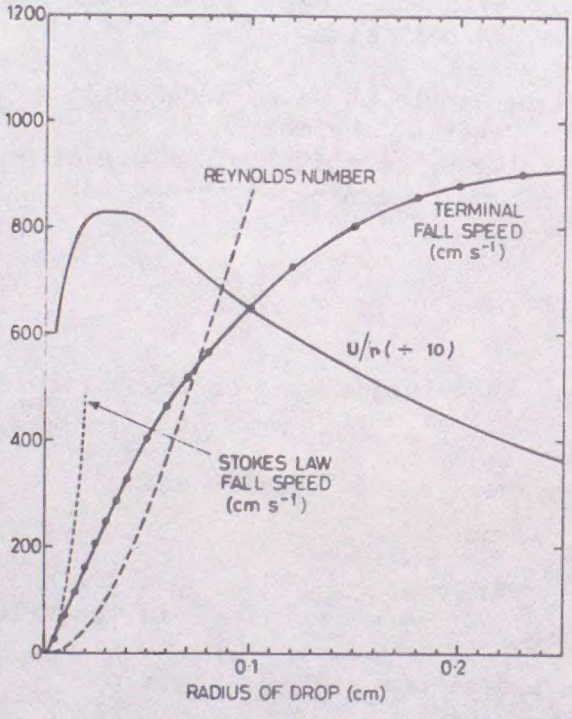
$$\rho_p a^2 > \frac{9\eta}{2}\frac{r}{u} \qquad (13)$$

Thus for a given aerosol the efficiency of removal is decided by the ratio of the drop radius to its terminal velocity. Those drops for which U/r is a maximum will capture the smallest aerosol.

For large cloud drops their terminal velocity follows Stokes law:

$$mg = 6\pi r\eta U \quad \text{or} \quad U/r = \frac{2\rho r g}{9\eta} \qquad (14)$$

As found in lecture (2), large cloud drops are more efficient collectors than small drops. Above r=40 μm the terminal velocity of a drop falls below the Stokes law prediction and eventually departs significantly from this as sphericity is lost.



Terminal fall speed, Reynolds Number and the ratio U/r (fall speed to radius) for precipitation-size water drops.

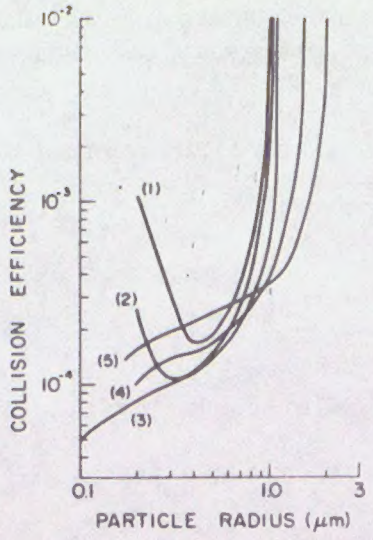
As shown above U/r reaches a maximum of 8300 sec^{-1} at about r=300 to 400 μm

Then $$\rho_p a^2 \geq \frac{9}{2}\eta . 1.2 10^{-4}$$

$$a \geq 2\mu m \text{ for } \rho_p \sim 2 g cm^{-3}.$$

Thus a rather abrupt transition from efficient collection to poor or no collection is predicted around a $\simeq$ few μm.

An exhaustive treatment of the problem has been made by Beard (1974) and Grover (1978). Some typical collection efficiencies calculated by the latter author are shown below.



Numerically computed efficiency with which water drops collide by inertial impaction with aerosol particles in air of 10 °C and 900 mb; (1) $N_{Re} = 200$ ($a = 438\ \mu$m), (2) $N_{Re} = 100$ ($a = 310\ \mu$m), (3) $N_{Re} = 30$ ($a = 173\ \mu$m), (4) $N_{Re} = 10$ ($a = 106\ \mu$m), (5) $N_{Re} = 4$ ($a = 72\ \mu$m). (From Grover, 1978).

A threshold at about a=1 μm is evident. The minimum and subsequent slight increase in collection efficiency of small particles by large drops (curves 1 and 2) is a result of their capture by a standing wake eddy which is predicted at the large Reynold's number appropriate to these drops.

Such collision efficiencies are related to the collection kernel, K(r,a) and scavenging coefficient, Λ introduced earlier, by:

$$K(r,a) = \pi (r+a)^2 E(r,a) \left[u_r - u_a \right] \quad (15)$$

2.4 Electrical effects

The atmosphere is ionised by the action of cosmic rays, natural radioactivity and electrical discharge processes in the vicinity of thunderstorms. Aerosol (including cloud drops etc) acquire charge through Brownian deposition of these ions. In turn such particles experience electrostatic attraction or repulsion as a result of that charge. It is instructive to consider whether such electrical forces can influence scavenging.

A rather simple approach considers the diffusion of charges of both sign to a Boltzmann equilibrium distribution on particles of radius a. The resulting net average charge is zero of course but the average number of elementary charges irrespective of sign is given by:

$$|\bar{p}| = 300\sqrt{a}$$

where a is in cm.

Thus at equilibrium a 5 μm radius drop is expected to capture, on average, rather less than 10 elementary charges. This represents very

weak electrification; the electric energy of the drop being orders of magnitude less than its kinetic energy at terminal velocity or its surface free energy.

If conduction under the action of an external field is added to the process of diffusion then a maximum charge of

$$Q_{max} = 3Er^2 \left\{ \frac{\left(\sqrt{\frac{\lambda_+}{\lambda_-}} - 1\right)}{\left(\sqrt{\frac{\lambda_+}{\lambda_-}} + 1\right)} \right\} \qquad \text{—— (16)}$$

where λ_+, λ_- are the polar conductivities of the atmosphere.

Away from ionic sources, such as corona sites, $\lambda_+/\lambda_- < 2$ so that

$$Q_{max} \doteq \frac{1}{2} E r^2 \qquad \text{—— (17)}$$

is a useful approximation.

Takahashi (1973) has summarised measurements of charge on cloud and precipitation particles for conditions of both strong and weak electric fields.

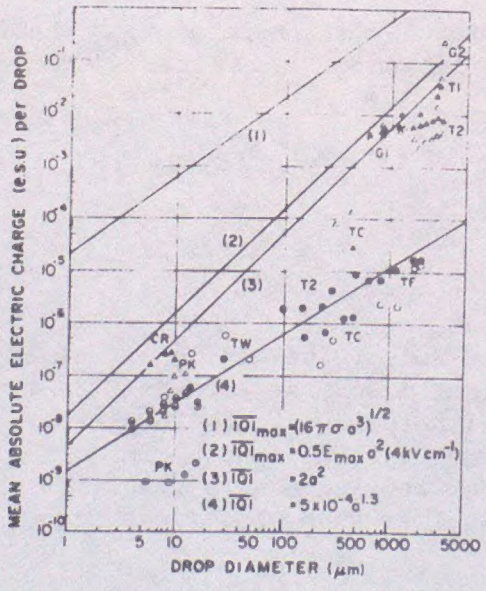


Fig. 17-2. Mean absolute electric charge on cloud and raindrops. Round symbols indicate warm cloud cases, triangular symbols indicate thunderstorm cases; solid symbols indicate negative charge, open symbols indicate positive charge. PK Phillips and Kinzer (1958), S Sergieva (1959), CR Colgate and Romero (1970), TW Twomey (1956), TC Takahashi and Craig (1973), T1 Takahashi (1965), T2 Takahashi (1972), TF Takahashi and Fullerton (1972), G1 Gunn (1949), G2 Gunn (1950).

Curve 1 defines the so called Rayleigh limit set by the equality of surface tension and electrostatic stress, ie. $E_{max}^2/8\pi = 2\sigma/r$.

As $E_{max} = Q_{max}/r^2$

$$Q_{max} = (16\pi\sigma r^3)^{1/2} \text{ esu} \qquad \text{—} \qquad (18)$$

Curve 3 represents an empirical relationship

$Q_{max} = 2r^2$ which is compatible with (17) above for $E \sim 1.2\,10^5\ V m^{-1}$, a typical figure for the field in a thunderstorm cloud.

Curve 4 is an empirical expression for the charge on weakly electrified drops. $Q = 0.2a^2$ finds common use also and is a fair fit to the data; from (17) the implied field is $\sim$100 × greater than the fair weather field nevertheless.

Even if the charges on a drop and aerosol particle are zero, induced dipole charges generated by an external field Eo produce an attractive force between them. These effects have been considered by Grover et al (1977) who model the efficiency with which water drops collect aerosol under the action of inertial, thermoporetic diffusiophoretic and electric forces.

The diagram shows the collection efficiency of a drop of radius 106 μm for aerosol particles of density 2g cm^{-3} and indicated radius. 2a,b,c for $q_a = \mp 0.2a^2$, $q_r = \pm 0.2r^2$ and RH=100%, 95% 75% respectively.
3a,b,c for $q_a = \mp 2.0a^2$, $q_r = \pm 2.0r^2$ and RH = 100%, 95%, 75% respectively.
The curves 2 are practically indistinguishable from those for which qa=qr=0, but the presence of charges characteristic of thunderstorms does have an appreciable effect.

2.5 Nucleation

As was shown in lecture (1), the condensation process is driven by the supersaturation of the air but a nucleus is necessary to initiate this process. The critical supersaturation for activation can be expressed as

$$S_c = \frac{8\sqrt{2\pi}}{9\rho_L}\left(\frac{\sigma M_w}{RT}\right)^{3/2}\left(i m \frac{M_w}{M_N}\right)^{1/2} \qquad \text{—} \qquad (19)$$

where M_w, M_N is the molecular weight of water and nucleus respectively
σ is the surface free energy of water
ρ_L is the density of water
m is the mass of the dry nucleus material
i is the van't Hoff factor - see lecture (1)

For NaCl ($i=2$, $M_n = 58.4$, $\rho_N = 2.17$ g cm^{-3})

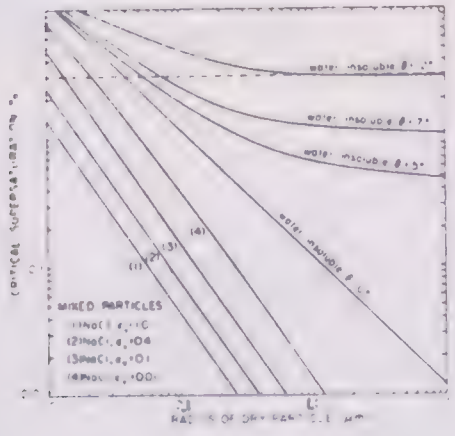
$$r_c = 1.16\,10^{-6}\,S_c^{-2/3}\ \text{cm}$$ where S_c is in per cent.

For $(NH_4)_2SO_4$ ($i=3, M_n=132.1$, $\rho_N = 1.77$ g cm^{-3})

$$r_c = 1.4\,10^{-6}\,S_c^{-2/3}$$

Thus for Sc=0.5% the critical dry radius is $\sim$ 0.02 μm for both NaCl and $(NH_4)_2SO_4$.

Such a dry nucleus will grow to almost 0.1 μm radius at 100%RH and once the critical supersaturation is reached will rapidly increase to a few microns. The diagram below from Pruppacher and Klett(1978) shows how the radius of a nucleus formed from a mixture of soluble and insoluble material (the latter being characterised by a contact angle in the absence of a soluble component) is related to the critical supersaturation.



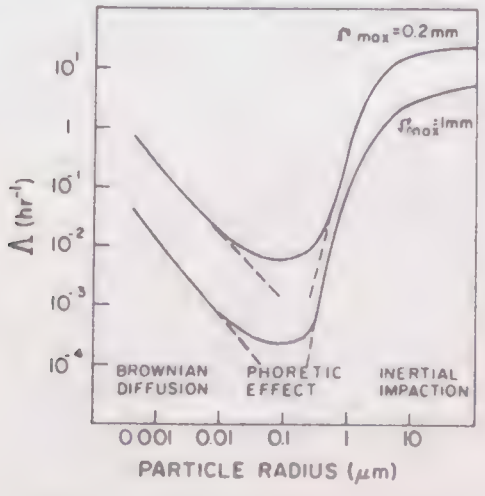
Obviously this process allows soluble aerosol in the 0.1 to 1.0 μm radius interval to become part of cloud drops.

3 Overall scavenging effect

3.1 Theoretical predictions

It is not obvious that the supersaturation of the individual effects described above are simply additive. The presence of phoretic effects modifies symmetrical diffusive processes and diffusion increases the effective cross-section for inertial capture.

Nevertheless the broad features, of effective scavenging of small aerosol particles by diffusion and of large particles by inertial capture are confirmed when the various processes are combined. Greenfield (1957) first considered Brownian diffusion and inertial impaction. He found that the overall scavenging coefficient exhibited a strong, broad minimum between a=0.1 μm and 1.0 μm - the so called 'Greenfield gap'. Later investigations, eg Slinn and Halen (1971), Pilat and Prem (1976) and Wang and Pruppacher (1978), who included contributions from diffusion, inertial capture and diffusio-and thermophoresis, have obtained similar results.



Scavenging coefficient as a function of aerosol particle size for Brownian diffusion, inertial capture, and thermo- and diffusiophoresis; $\Delta T = T_\infty - T = 3\,°C$, precipitation rate $R = 10\,mm\,hr^{-1}$; raindrop size distribution $n(r)\,dr = (10^{-4} R/6\pi r_{max}^7) a^2 \exp(-2r/r_{max})\,dr$, with R in $cm\,sec^{-1}$; drop terminal velocity $V_\infty = 8000\,r$ (sec^{-1}) with r in cm, collision efficiency for inertial impaction based on values of Zimin (1964). (From Slinn and Hales, 1971)

More recently Grover et al (1977) and Wang et al (1978) have added electrical effects and probably achieved the greatest rigour in their analysis. Their results in terms of collection efficiency are shown below.

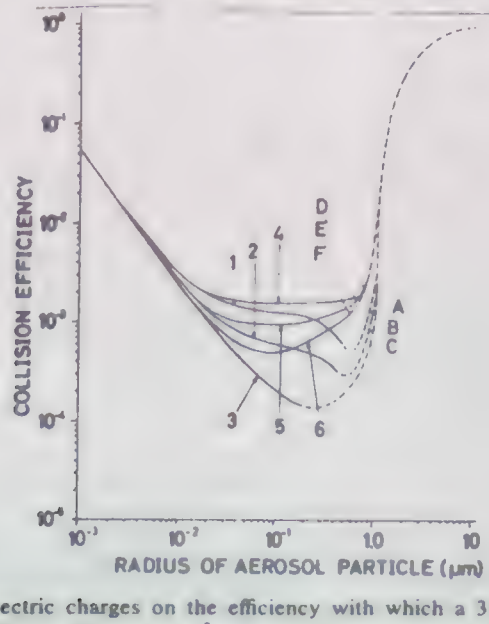


Fig. 17-10. Effect of electric charges on the efficiency with which a 310 μm radius water drop collides with aerosol particles of $\rho_p = 2\,g\,cm^{-3}$ in air of various relative humidities ϕ_v, and of 10 °C and 900 mb. Composite results combining the particle trajectory method of Grover *et al.* (1977) (dashed lines) with the particle flux model of Wang (1978), and Wang and Pruppacher (1978) (solid lines). Curves 1, 2, 3, i.e. A, B, C are for $\hat{Q}_a = 0$, $\hat{Q}_r = 0$ and $\phi_v = 50\%$, 75%, and 95%, respectively. Curves 4, 5, 6, i.e. D, E, F are for $\hat{Q}_a = \pm 2.0$, $\hat{Q}_r = \mp 2.0$ and $\phi_v = 50\%$, 75%, and 95%, respectively; where $\hat{Q}_a = Q_a/a^2$ and $\hat{Q}_r = Q_r/r^2$. (From Wang, 1978, with changes.)

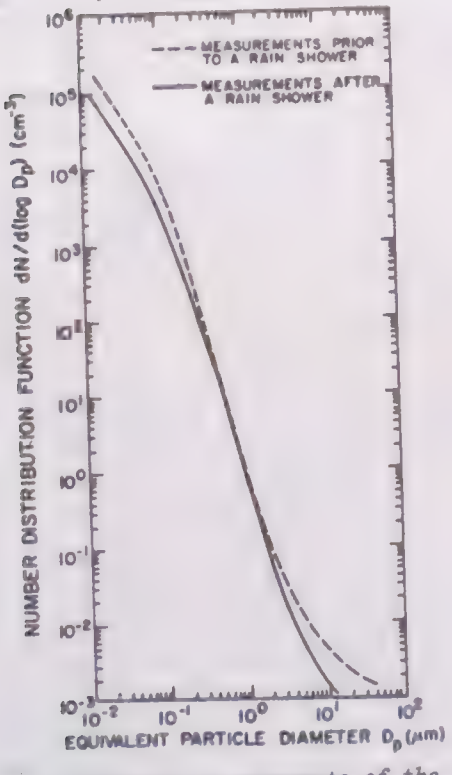
The phoretic processes tend to fill the gap but it remains distinct nonetheless.

The atmosphere does provide a mechanism to 'bridge the Greenfield gap' in that the soluble particles of radii between 0.1 and 1.0 μm are those which most readily serve as cloud nuclei. Of course this mechanism is not open to insoluble particles.

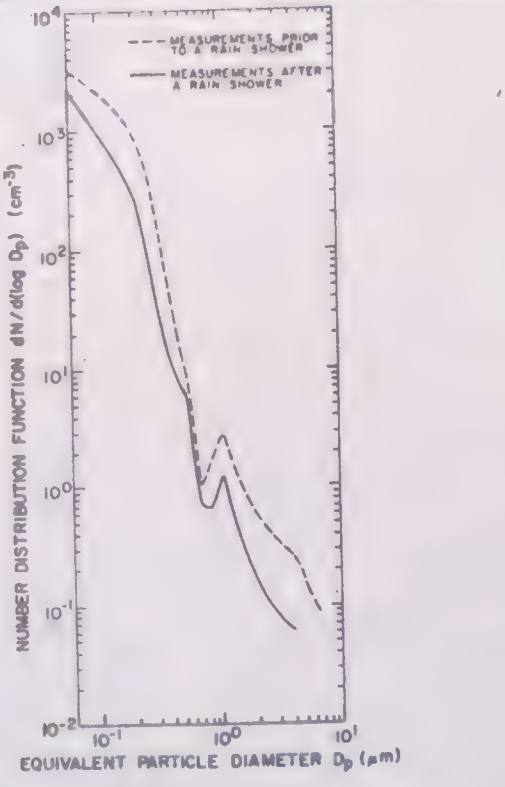
Experimental results

So far as is known only the University of Washington group, Radke et al (1974), Hobbs et al (1978) and Radke et al (1978) have published atmospheric measurements of the detailed effects of precipitation scavenging. Most of their data have been obtained in particulate plumes just before and just after the plumes were scavenged by rain or snow showers. Obviously the experiment is a difficult one to engineer!

Examples of two data sets are shown below.



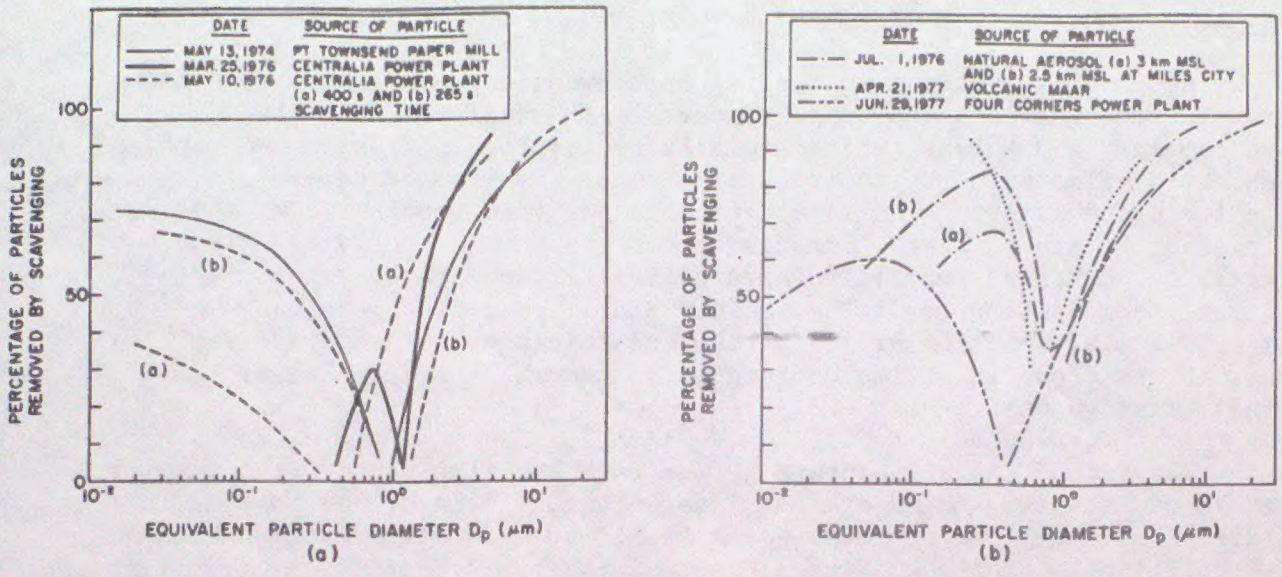
Airborne measurements of the size spectra of particles in the plume from the coal-fired electric power plant at Centralia, Wa. measured at 5 km (12 min. travel time) downwind of the stack on May 10, 1976.



Airborne measurements of the size spectra of particles at 2.5 km MSL near Miles City, Mt. on July 1, 1976.

Although the particle size spectra for the two cases are quite different their responses are similar. Both show minimal differences between the scavenged and unscavenged samples in a narrow size region around 1 μm diameter and both show marked decreases due to scavenging for particles smaller and greater than 1 μm. The percentage of

particles of various sizes removed by precipitation are shown below for a number of case studies.



The upper edge of the 'Greenfield Gap' is in good agreement with theory but at smaller sizes the measurements are in disagreement with theory. The authors suggest that the apparent high removal efficiency for particles in the vicinity of d=0.1 μm may be because they had a high hygroscopic component and were therefore of greater size than the 'dry' size shown. (Their instrumentation dries the air sample before measurement).

Wang and Pruppacher (1977) have reviewed earlier attempts to define collection efficiency in the laboratory and published results in excellent agreement with theory.

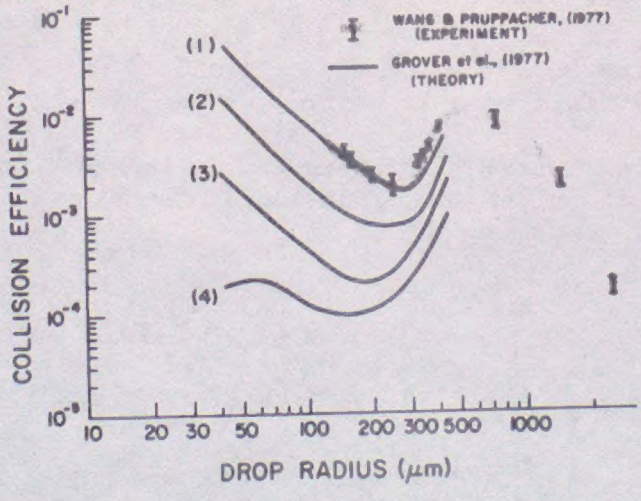


Fig. 12-9. Variation of the efficiency with which water drops collide with aerosol particles of $\bar{a}$= (0.25 ± 0.03) μm, ρ_p = 1.5 g cm^{-3} in air of 1000 mb, as a function of drop size and relative humidity ϕ_v of the air. Experimental results are due to inertial forces, phoretic forces, and Brownian diffusion, in air of ϕ_v = 23%, T = 22°C, and p = 1000 mb. Theory considers inertial and phoretic effects only, (for such a particle size the collection efficiency due to Brownian diffusion is about one order of magnitude lower than that due to phoretic effects – ; (1) ϕ_v = 20%, (2) ϕ_v = 75%, (3) ϕ_v = 95%, (4) ϕ_v = 100%. (From Wang and Pruppacher)

The maximum at r $\sim$ 500 μm is a further manifestation of the wake capture of sufficiently small aerosol. At drop sizes above 500 μm these eddies are progressively shed. The low relative humidity (and consequent drop evapoaration) generated strong phoretic effects in the experiment, which are of limited applicability to the atmosphere.

Summary

Apparently in-cloud scavenging by convective, precipitating systems may be regarded as a two stage process. In the first, all the scavenging mechanisms, and especially nucleation, diffusion and perhaps phoretic effects, serve to transfer a major fraction of aerosol, a $\leq$ 1 μm, to the cloud water. The time available for this process is of the order of 20 min or less. In the second stage this 'polluted' cloud water is scavenged primarily through inertial capture by relatively large precipitating particles. This accretion process can cause a substantial fraction of the aerosol containing cloud water to fall out of the cloud in a time comparable to, though generally larger than, that of stage one.

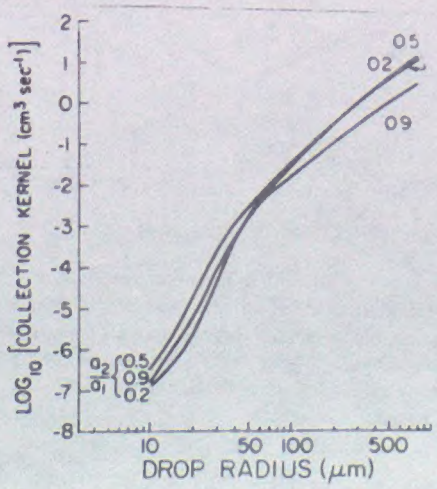
The second stage is generally the rate limiting one. Conceptually at least, the two stages are not independent. Thus clouds forming in 'dirty' air may lock up their water in a drop size distribution which is colloidally stable and therefore an inefficient producer of precipitation. Rather little is known about this in a quantitative sense. Nevertheless a reasonable upper bound for the effective scavenging rate is given by the accretion rate of small cloud particles by precipitation, with the former assumed to have absorbed most of the aerosol particle mass through the stage one process. Such a formulation also represents an upper bound for the case of below cloud scavenging (wash-out) - perhaps an unrealistic one as inertial capture is efficient for the large particle sizes only. However Radke et al (1978)'s results may provide a clue to an effective short circuit for this.

Thus following Pruppacher and Klett (1978) if a steady state rain spectrum - see lecture (2) - is assumed then

$$n(r) = n_o (\exp(-\lambda r) \qquad (20)$$

where r is in cm, $n_o = 1.6\ 10^5\ m^{-3}\ cm^{-1}$ and $\lambda = 82\ R^{-0.21}\ cm^{-1}$ where R is the rainfall rate in mm hr^{-1}.

The diagram below taken from Long (1974) suggests that the collection kernel is only a weak function of the size of the collected drop.



Collection kernel $K(a_1, a_2)$ based on collision efficiencies of Shafrir and Gal-Chen (1971), and Klett and Davis (1973). (From Long, 1974)

Thus, equations (9), (15) and (20) allow specification of

$$\Lambda_{max} = \pi n_o \bar{E} \int_0^\infty r^2 U(r) e^{-\lambda r} dr$$

For these purposes it is possible to use $U(r) = 3200 r$ cms^{-1}
$\bar{E}$ represents a characteristic collection efficiency for drop/drop interactions, where $E=O(1)$ but rather less than this for aerosol particles < 1 μm - see section 2 and 3.

Then, bearing in mind that a number of assumptions have been made to maximise the scavenging rate,

$$\Lambda_{max} = 1.5 \bar{E} R^{0.79} \text{ hr}^{-1}$$

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